Supporting Information

Highly safe and stable lithium-metal batteries based on a quasi-solid-state electrolyte†

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Supplementary Figures

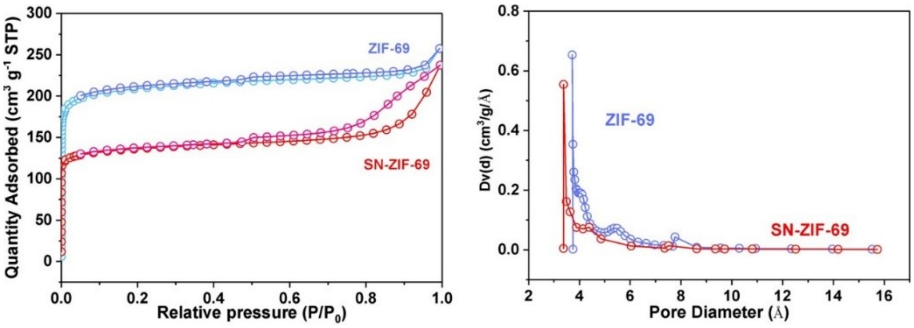

Figure S1. (a) N_2 adsorption/desorption isotherms of and (b) pore size distribution of ZIF-69 and prepared SN-ZIF-69 electrolyte.

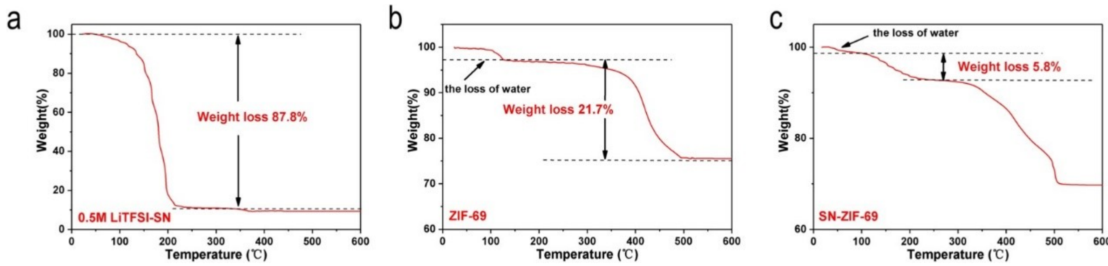

Figure S2. TGA curve of (a) 0.5M LiTFSI-SN electrolyte, (b) ZIF-69 material, and (c) prepared SN-ZIF-69 electrolyte. It can be clearly observed that the ZIF-69 possesses a certain thermal stability below 300°C. The ~3 wt% weight loss for ZIF-69 up to ~100 °C corresponds to the removal of adsorbed water. In TG result of 0.5M LiTFSI-SN electrolyte, the SN solvent will be entirely removed by the temperature elevated to ~200°C. Finally, in TG result of SN-ZIF-69 electrolyte, the ~5.8 wt% weight loss for SN-ZIF-69 up to ~200 °C corresponds to the removal of SN solvent from ZIF-69, indicating that the loading mass of SN-based electrolyte in SN-ZIF-69 electrolyte structure is ~5.8 wt% (The solvent-depleted SN-based electrolyte was assumed to load in ZIF-69 structure).

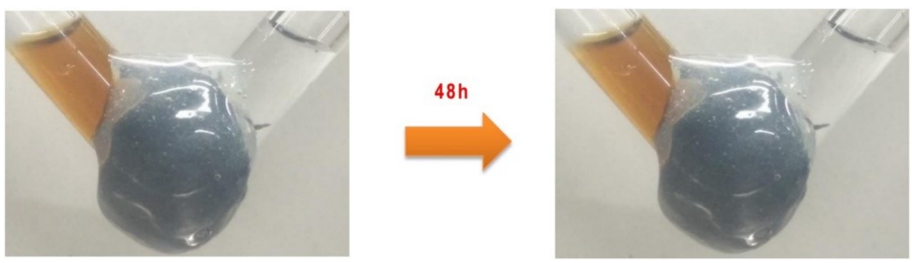

Figure S3. Diffusion experiment of molecules with large size (polysulfides, 9 Å) employed to verify the excellent compactness of the obtained ZIF-69 film.

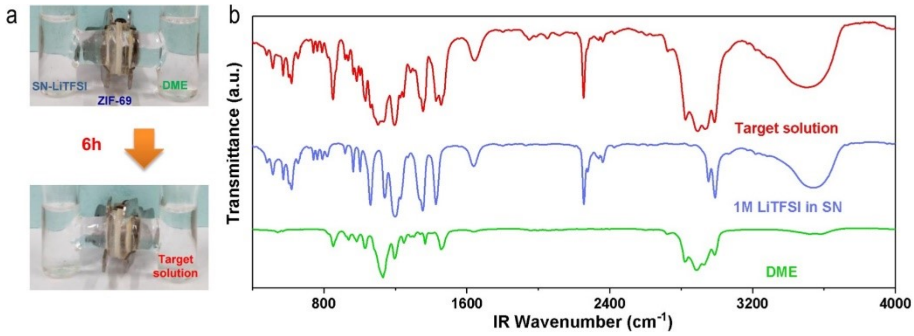

Figure S4. (a) Diffusion experiment of small molecules (SN solvent molecule) and (b) its corresponding FT-IR spectra of the marked solution employed to identify its capability to transport molecules with specific sizes.

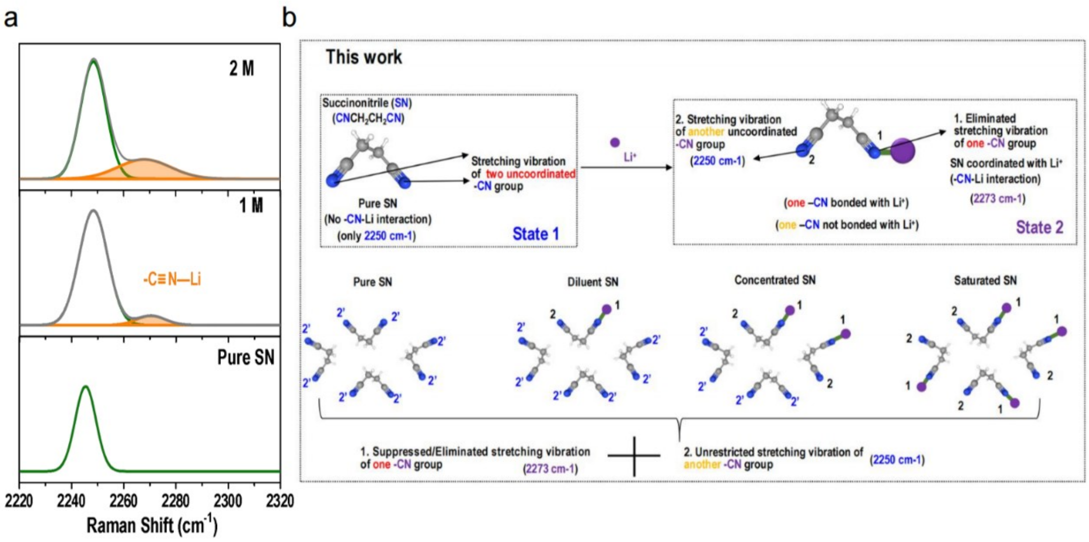

Figure S5. (a) Raman spectra of pure SN solvent, 1M LiTFSI-SN electrolyte, and 2M LiTFSI-SN electrolyte to demonstrate the gradually increased -C≡N-Li-related signal by the rising LiTFSI salt concentration in SN solvent. (b) The Coordination states of SN with Li^+ within electrolytes of different concentrations in our work.

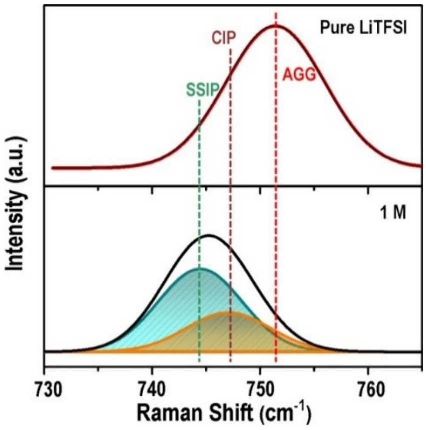

Figure S6. Raman spectra of pure LiTFSI salt, and 1M LiTFSI-SN electrolyte.

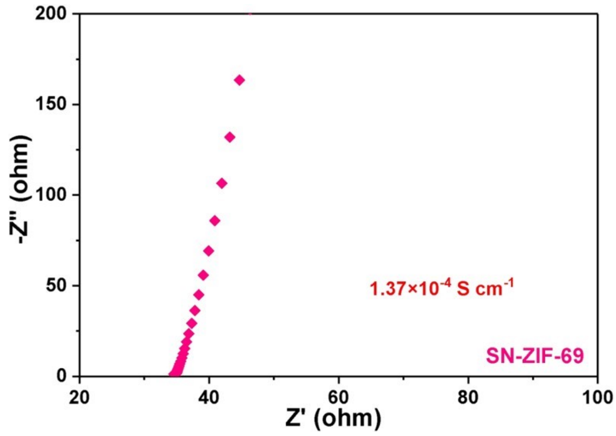

Figure S7. Nyquist plot of the SS/SN-ZIF-69 electrolyte/SS at room temperature.

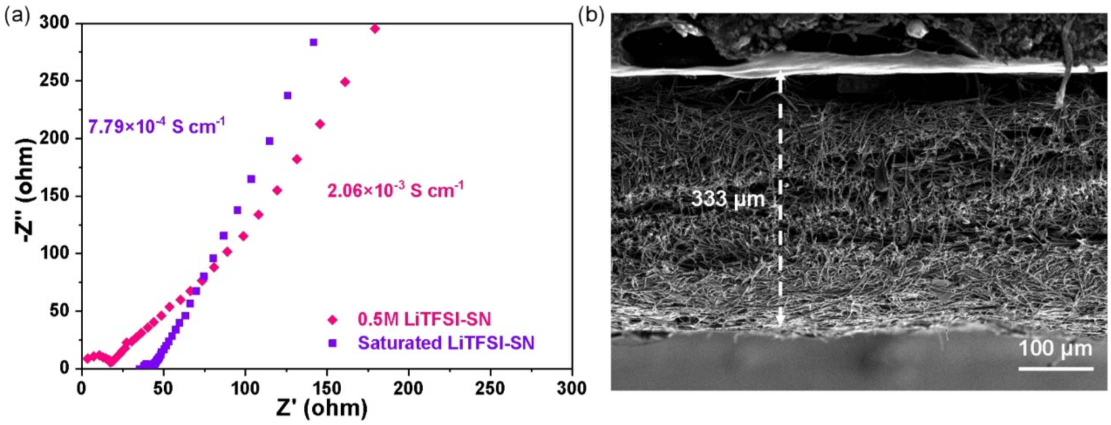

Figure S8. (a) The Nyquist plot of the SS/0.5M LiTFSI-SN electrolyte/SS and SS/Saturated LiTFSI-SN electrolyte/SS at room temperature. (b) The SEM image of glass fiber applied in cell assembling to show the thickness of separator.

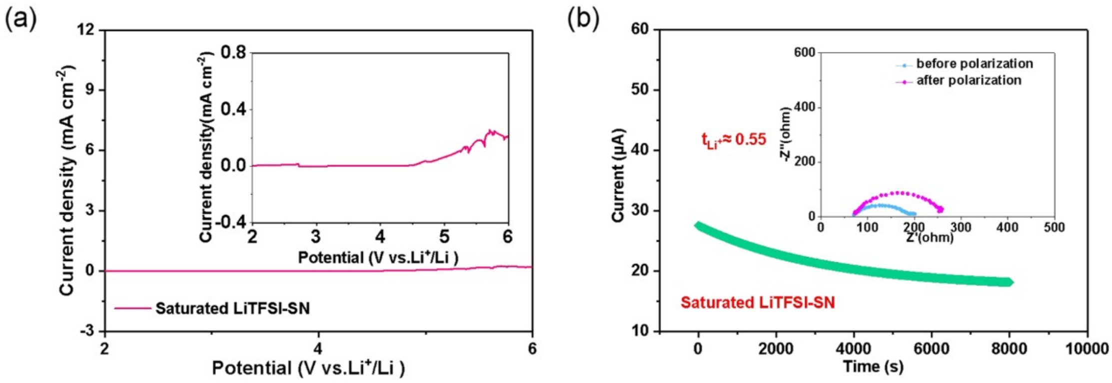

Figure S9. (a) Linear sweep voltammetry curves (LSV) of the saturated LiTFSI-SN electrolyte. The inset is the enlarge version. (b) Polarization curves the electrochemical impedance spectra (inset) of the saturated LiTFSI-SN electrolyte used to calculate the lithium-ion transference number (t_{Li^+}).

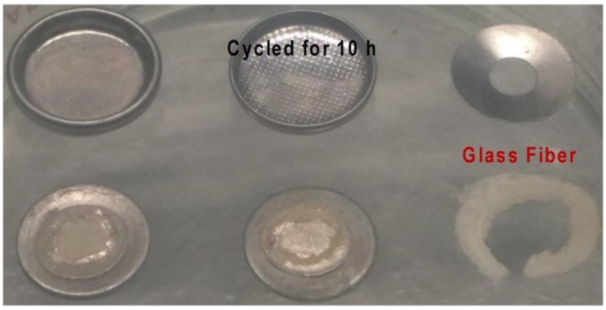

Figure S10. Lithium metal anodes and corresponding electrolyte films collected from lithium symmetrical batteries equipped with 0.5M LiTFSI-SN electrolyte after 10 hours cycling. The visible colored nitrile polymerization products on the lithium surface can be verified.

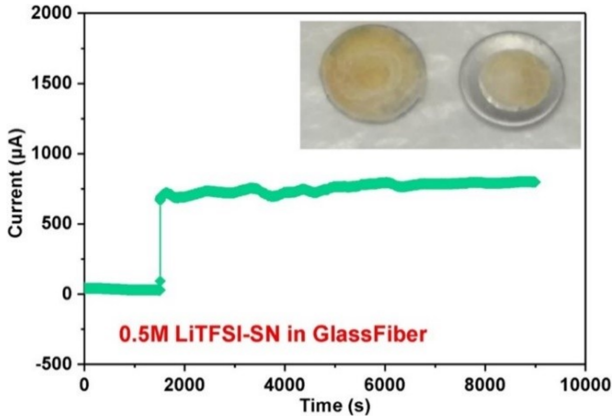

Figure S11. Polarization curves of the 0.5M LiTFSI-SN electrolyte. The inset shows the corresponding lithium anode and glass fiber separator collected from disassembled battery after chronoamperometry test. The uncontainable nitrile polymerization on lithium surface may cause the abnormal polarization curves.

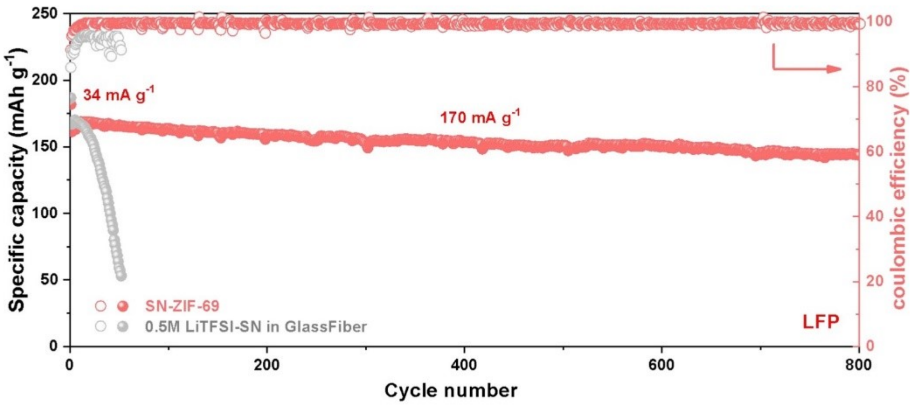

Figure S12. Cycling performance of the LFP//Li half-cell used the SN-ZIF-69 electrolyte (red) and 0.5M LiTFSI-SN electrolyte (grey).

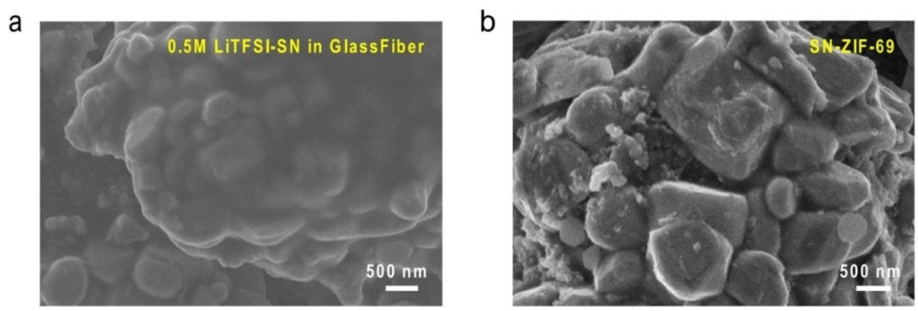

Figure S13. SEM images of the cycled NCM-111 cathodes collected from different batteries equipped with (a) 0.5M LiTFSI-SN electrolyte, and (b) SN-ZIF-69 electrolyte after 50 cycles.

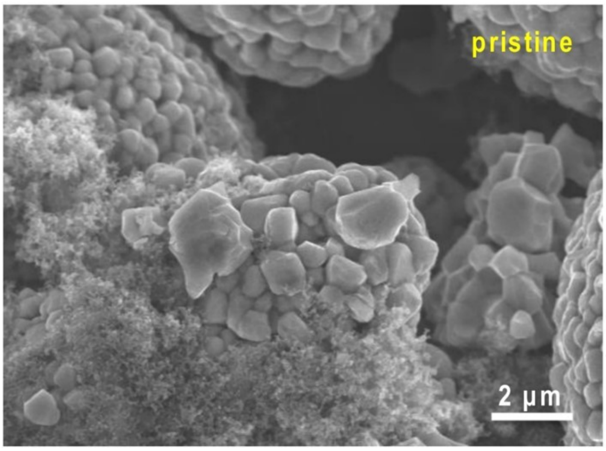

Figure S14. SEM images of pristine NCM-111 cathode.

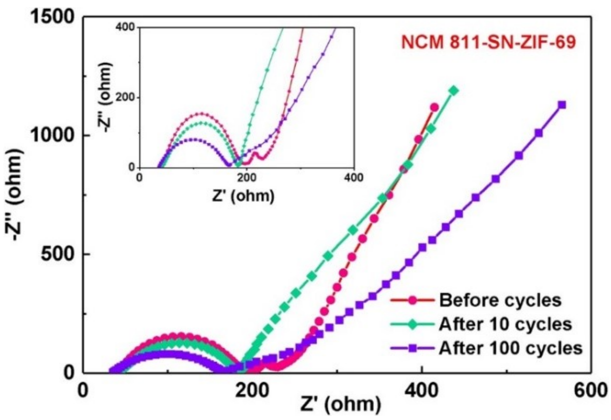

Figure S15. Nyquist plots of the NCM-811/SN-ZIF-69 electrolyte/Li before and after several cycles at room temperature.

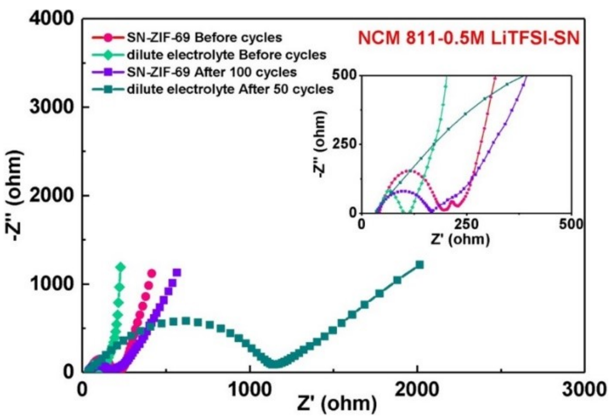

Figure S16. Nyquist plots of the NCM-811/SN-ZIF-69 electrolyte/Li and NCM-811/0.5M LiTFSI-SN electrolyte/Li before and after 100 cycles at room temperature.

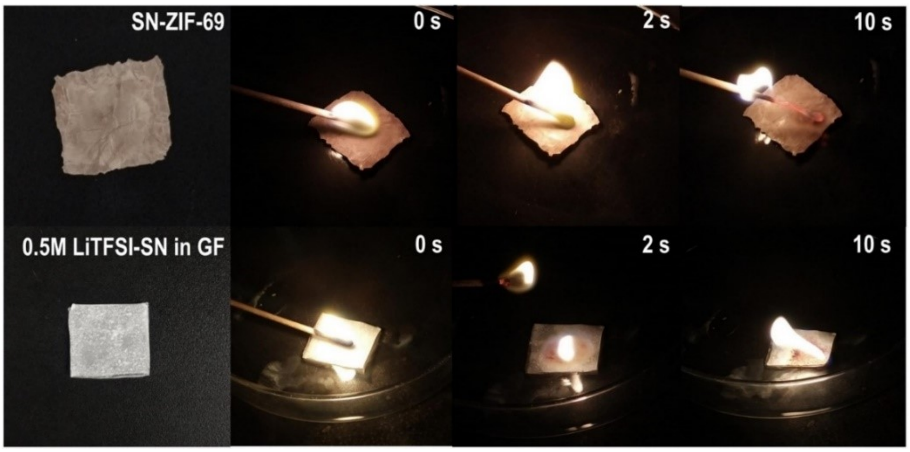

Figure S17. Combustion tests of the glass microfiber filter dipped with 0.5M LiTFSI-SN electrolyte and prepared SN-ZIF-69 electrolyte.

Figure S18. Lithium metal pouch-cell equipped with SN-ZIF-69 electrolyte and LFP cathode.

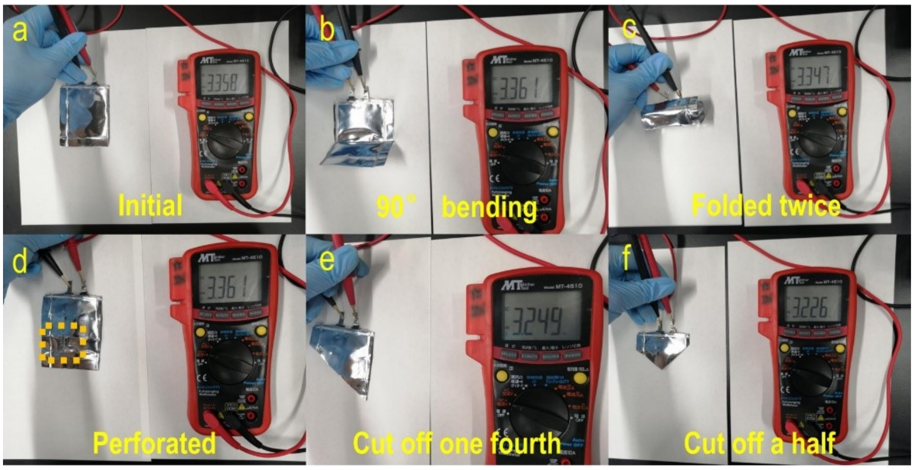

Figure S19. The OCV test of as-obtained LFP/ SN-ZIF-69/Li pouch-cell under various harsh conditions such as cutting, folding and perforation.

Comparative result of electrochemical performance				
evaluating indicator Work classification	Cathode system	Upper limit of charging voltage	Cycle number	Reversible capacity (capacity retention)
Our work (SN-ZIF-69 electrolyte)	NCM-811 (cathode)	4.6 V	200	164.5 mAh g^{-1} (84.2%)
Ref. 22a (SN-5%FEC)	Sn electrode (anode)	2 V	400	265 mAh g^{-1} (~60.2%)
Ref. 22b (5 mol% LiTFSI/SN in cathode)	C6Q (organic cathode)	3.7 V	500	405 mAh g^{-1} (~95.3%)
Ref. 22c (SN-LiTFSI-LiODFB-10%FEC)	LCO (cathode)	4.4 V	200	~130 mAh g^{-1} (85%)
Ref. 22d (SN-20%FEC)	LCO (cathode)	4.2 V	100	~124 mAh g^{-1} (93%)
Ref. 22e (SN-5%FEC)	LFP (cathode)	4.2 V	1000	~110 mAh g^{-1} (81%)

Table S1. Comparative result of electrochemical performance between our work and other's work.

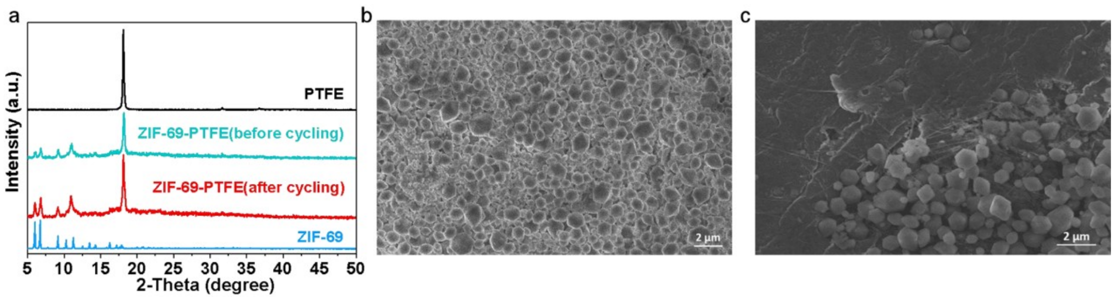

Figure S20. (a) XRD patterns of PTFE, ZIF-69, ZIF-69 film before and after electrochemical cycling. SEM images of (b) prepared ZIF-69 film and (c) residual ZIF-69 film on a 300 hours-cycled cycled lithium anode surface.

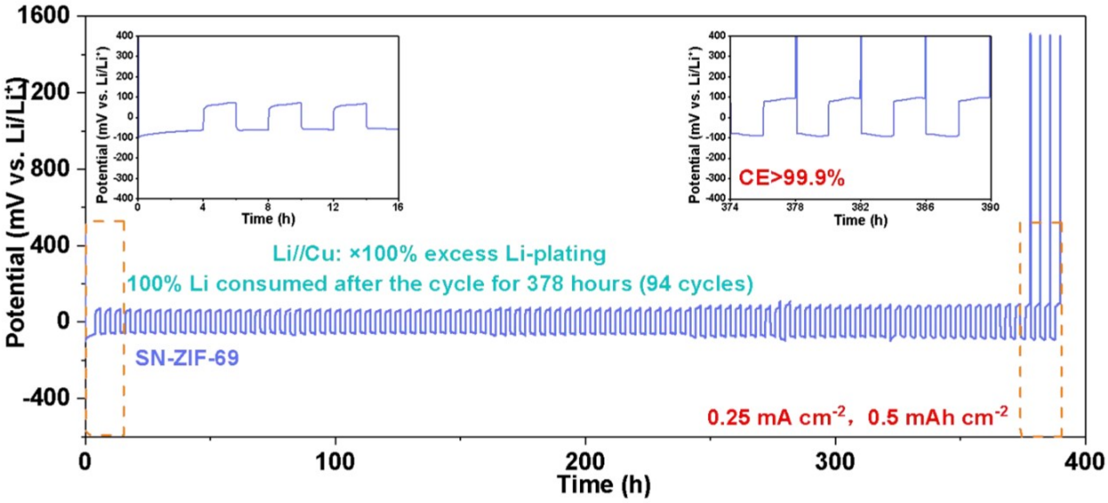

Figure S21. The cycle performance of Li //Cu half-cell (with one-time excessed Li pro-deposited on copper) equipped with SN-ZIF-69 electrolyte.